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EFFECT OF CAVITATION ON HIGH-TEMPERATURE SILICATE MELTS

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The effect of acoustic vibrations in the cavitation regime on high-temperature high-viscosity slag melt is evaluated experimentally. It is determined that ultrasonic treatment greatly increases the crystallization power of melt; this could be due to depolymerization of the silicon-oxygen framework of silicate melt and the creation of an extended interface between the gaseous and liquid phases in the melt. It is shown that acoustic treatment promotes dispersion of a refractory solid phase in the melt.

Key words: acoustic vibrations, cavitation, silicate melt, crystallization, mixing, waveguide.

Mixing and production of uniform melts play an important role in the manufacture of articles made of glass and glass ceramic materials at all stages of glassmaking. Conventionally, mechanical mixers, bubbling and electric heating are used to mix glass melts; these operations intensify convective flows in molten glass. Acoustic treatment of melt in the cavitation regime can have an intensifying effect on glass formation, homogenization and fining of molten glass.

The acoustic treatment of liquid media is used quite widely in chemical technology to intensify different processes [1]. In the primary method high-intensity acoustic vibrations are introduced directly into a liquid medium by immersing the working tool of the vibratory system (waveguide) into the process volume of the liquid. For treatment in the cavitation regime, cavitation steam-gas bubbles arise in the medium. On expansion these bubbles accumulate energy and under compression they can explode with intense emission of acoustic shock waves producing strong hydrodynamic disturbances in the liquid — intense mixing, alternation motion of particles and intensification of mass-transfer processes. In a number of cases structural changes, for example, mechanical destruction of polymers, are observed in the medium [2].

Conventionally, these technologies are used to intensify dissolution, purification, extraction and dispersion processes in low-viscosity aqueous media and organic solvents. In recent years experiments on ultrasonic intensification of processes have also been conducted in high-viscosity media as well as in media with a high concentration of a solid phase [3].

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In the present study we use a model experiment to evaluate the effect of cavitation on high-temperature silicate melt, determine the possibility of mixing a refractory solid phase in a viscous silicate melt and check the applicability of a molybdenum waveguide for creating acoustic oscillations in melt at temperatures characteristic for glassmaking.

The experimental object is blast furnace slag with the following composition (wt.%): 44.30 SiO_2; 5.48 Al_2O_3; 44.40 CaO; 4.59 MgO; 0.44 Fe_2O_3; 0.44 SO_3, forming a uniform melt at 1450°C. The viscosity of the melt at this temperature is approximately 10 Pa · sec. Glassy quartz sand with 0.3 – 0.5 mm grains was used as the dispersed solid phase.

The experiment was performed on apparatus consisting of a high-temperature laboratory electric furnace with silicon carbide heaters and a platinum – platinum-rhodium thermocouple, an ultrasound generator with variable power to 5 kV (developed and manufactured by Kriamid, JSC) and a cylindrical molybdenum waveguide-radiator (Fig. 1). The diameter of the waveguide was 40 mm, the gain 1, the maximum amplitude of the end-face vibrations 15 μm, and the oscillatory frequency 17.4 kHz. The waveguide was designed for and tuned to the resonance frequency of the PMS15A-18 transducer taking account of the fact that it will operate in melt at 1450 – 1500°C. The waveguide surface is coated with a liquid glass solution to decrease corrosion of molybdenum in air.

To perform the experiments 250 g of blast-furnace slag in a 700 ml corundum crucible were placed in a furnace, a waveguide was inserted into the crucible from the top and the furnace was heated to 1450°C. After the slag melted the waveguide was found to be immersed in the melt to depth about 50 mm. After the temperature in the furnace stabilized

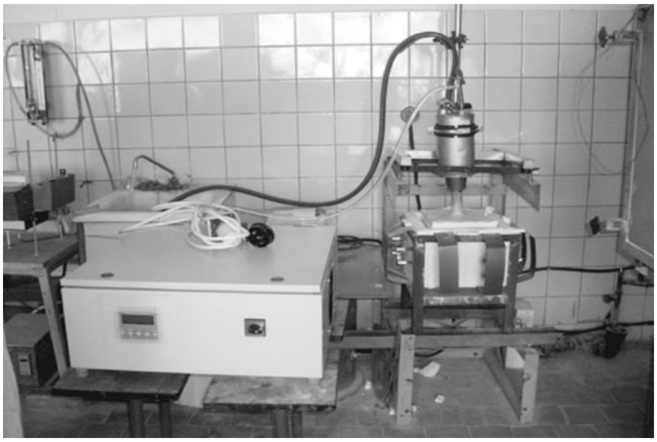

Fig. 1. Experimental setup.

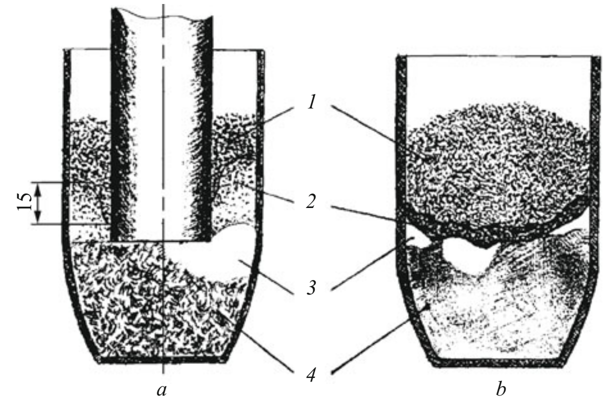

Fig. 2. Visual appearance of the material in crucibles after completion of the experiments: *a*) with acoustic treatment; *b*) without acoustic treatment; *1*) quartz sand; *2*) slag layer with distributed sand grains; *3*) cavity; *4*) slag.

the acoustic generator was switched on. The experiment was conducted at the maximum amplitude of the oscillations and maximum power, which should generate cavitation in the slag melt. The melt treatment time was 1 h. At the completion of the experiment the crucible with the melt was cooled in the furnace to room temperature, removed from the furnace and cut apart with a diamond cutting wheel. In the control experiment the same method was used to obtain and heat-treat the slag melt but acoustic treatment was not used.

A comparative analysis of the material which did and did not undergo acoustic treatment shows that such treatment has a large effect on the structure of the slag melt; this manifested as a large increase of the crystallization power of the melt after such treatment. The untreated slag melt after cooling maintained a glassy state; there were no indications of crystallization. The treated slag melt partially crystallized on cooling, acquiring a coarse-crystalline structure. According to the results of x-ray phase analysis the acoustic action promoted crystallization of calcium aluminum-silicate — helenite $3CaO \cdot Al_2O_3 \cdot 2SiO_2$, apparently, with partial replacement of calcium and aluminum ions with magnesium and iron ions.

We note that the slag used is neutral with respect to the ratio of acidic and basic components, i.e., it is quite stable against crystallization. The higher proneness of the melt to undergo crystallization after acoustic treatment can be explained in two ways. One likely reason is depolymerization of the structural silicon-oxygen framework of the slag melt under cavitation, which greatly facilitates diffusion processes which promote crystallization of the melt. The rupture of chemical bonds with depolymerization of complex molecules under the action of high-intensity oscillations is known in chemical technology [4].

The second reason that the slag melt is more prone to crystallization could be the formation of cavitation voids due to the acoustic action that become filled with gas released from the melt. These voids create an extended interface between phases on which nucleation and growth of crystals of a silicate phase start.

To determine the possibility of the solid refractory phase being dispersed in high-viscosity slag melt under acoustic action 250 g quartz sand were added into the crucible, heated to 1450°C, with the melt on top, after which the vibration generator was switched on. The experimental procedure was otherwise unchanged. A section of the crucibles after they cooled is presented in Fig. 2.

Different zones are clearly visible in a section of the crucibles. A layer of quartz sand was found in the upper part of the crucible which had not undergone acoustic treatment. Immediately beneath it is a slag crust containing quartz sand particles; the crust is no than 5 mm thick. A void probably formed by gases released from the slag melt during heating, which because of the high viscosity of the slag crust with sand cannot escape into the atmosphere, is present at an even lower level. A solidified glassy layer formed by solidified slag melt is present at the bottom of the crucible. The same zones are present in the crucible where the melt underwent acoustic treatment, but the contact zone between the sand and slag melt is more extended. The sand in this zone is distributed to depth 15 mm, which confirms the intensifying action of cavitation on the mixing of refractory particles in high-viscosity silicate melt.

Picking the waveguide material which comes into contact with the corrosive melt is a separate problem of the acoustic treatment of high-temperature silicate melts. This material must meet a number of requirements, whose satisfaction ensures that the waveguide will work satisfactorily without breaking down under extreme operating conditions:

– adequate mechanical strength, making it possible to withstand mechanical loads which produce variable high-frequency deformations of the waveguide;

– heat tolerance, making it possible for the waveguide to operate at temperature to 1500°C;

– thermal stability, making it possible for the material to withstand temperature differences between the lower part of the waveguide, located in the melt at temperatures

1400 – 1500°C, and the upper part, located outside the furnace at ambient temperature;

– corrosion resistance at operating temperature to the oxidative atmosphere of air and to corrosive silicate melts.

In the present work molybdenum was chosen for the waveguide which must function in a high-temperature silicate melt at temperatures 1450 – 1500°C. After the experiment the lower part of the waveguide, immersed in the melt, did not show any visible changes, but the upper part was covered by a loose layer of shiny yellow crystals comprising molybdenum oxide MoO_3 even though the coating of solidified liquid glass was preserved. This attests to quite intense oxidation of the molybdenum by the oxygen in air at elevated temperatures, even though an attempt was made to protect it with liquid glass.

Protection of the surface of the molybdenum waveguide by dense coatings which are impermeable to air does not solve the problem, because during operation the geometric dimensions of the waveguide change as the guide compresses and expands with a high frequency (in the present experiment — 17.4 kHz). The dense and strong coating cannot withstand such deformations. One possible solution of the problem is to use water-cooled protective jackets or to fabricate the waveguide from special refractory ceramic materials.

In summary, it was established experimentally that acoustic treatment makes silicate melts more prone toward crystallization as they cool and intensifies mixing of the refractory particles in a viscous silicate melt. Aside from this it can be asserted that molybdenum can be used for the waveguide for research work: molybdenum is capable of withstanding a number of heat-treatments while remaining serviceable. At the same time thermal corrosion of this material precludes its industrial and experimental-commercial use in this capacity.

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